Eco-friendly synthesis of Zinc Oxide Nanoparticles using *Camellia sinensis* (Green Tea) and its characterization

Abstract:

Green synthesis is a simple and eco-friendly method of creating nanoparticles without toxic byproducts. Green tea, which is rich in polyphenols and flavonoids, can be used to synthesize ZnO nanoparticles. These nanoparticles have potential applications in medicine and various industries. The phenolic compounds in green tea act as effective reducers of metal ions, which stabilize nanoparticle growth. In this study, ZnO nanoparticles were synthesized using green tea leaves and characterized using various instruments. The Synthesized particle had an average particle diameter of 168.8 nm and a zeta potential of 38.2 mV. The UV-Vis analysis showed a blue-shifted absorption maximum at 323 nm, indicating that the synthesis of ZnOparticles ison a nano-scale. XRD spectra of calcined ZnO nanoparticles showed prominent diffraction peaks at various angles. Thermogravimetric analysis revealed that the degradation of the nanoparticles occurred rapidly at 657°C, and weight loss began at 753°C which confirms the thermal stability of ZnO nanoparticles.

Comment [Kw1]: Please mention a few.

Keywords: Zinc oxide,Polyphenols, Nanoparticles,TGA,TEM

Introduction:

Materials that are smaller than 100 nm are known as nanomaterials, and they exhibit atom-like behaviours due to their large surface area and wider band gap. Transition metal oxides and semiconductors that fall within the nanometer range have piqued interest in various fields(Dhanemozhi, Rajeswari, & Sathyajothi, 2017).Green synthesis is a method that reduces pollution and also offers several benefits, including simplicity, low cost, stable nanoparticles, quick production time, non-toxic byproducts, and the ability to synthesize on a large scale. There is an increasing demand for eco-friendly methods that do not involve the use of toxic materials in the synthesis process.

Green tea is abundant in polyphenols, particularly catechins, and other flavonoids. It is prepared without fermentation to prevent oxidation of these compounds. The composition of green tea is similar to that of a fresh leaf, with

some changes occurs during drying. Green tea contains phenolic compounds, proteins, amino acids, carbohydrates, lipids, and vitamins C and E(Cabrera, Artacho, & Gimenez, 2006). Phenolic compounds possess significant antioxidant potential and act as effective reducers of metal ions, making them ideal for supporting the eco-friendly synthesis of nanoparticles. Furthermore, the high level of proteins, lipids, and amino acids contribute to stabilizing nanoparticle growth and preventing particle agglomeration. ZnO NPs are metal nanoparticles with impressive properties, such as wide band gap, high piezoelectric property, and large binding energy. They are safe, non-toxic, and biocompatible, making them useful in various industries, including optoelectronics, diagnosis, and environmental protection.ZnONPs can have potential applications in medicine as anti-angiogenesis, anti-inflammatory, and anticancer agents. Their surface is rich in -OH groups, allowing them to dissolve in both acidic and strong basic conditions(Rajeshkumar, Agarwal, Venkat Kumar, & Lakshmi, 2018).

This study focused on synthesis of ZnO nanoparticles using green tea leaves and subsequently characterize them using Particle Size Analyzer (Dynamic light scattering), Ultravioletvisible spectroscopy, X-ray diffraction, Thermogravimetric analysis, and Transmission electron microscopy.

Materials and Methods:

The precursor Zinc sulphate heptahydrate with 99% purity and Molecular weight 287.56 g/mol was obtained from Himedia. Distilled water was used throughout the experiment. Sodium Hydroxide (NaOH), was used as a stabilizing agent. Loose green tea leaves were obtained from Mighty Bio-Nature Products, Coimbatore.

Green tea extract preparation:

About 10 g of green tea leaves were weighed and made into fine powder. Then it was dissolved in 100 ml distilled water. It was boiled in a water bath at 65^0C for 3 hours. Then it was filtered using Whatman filter paper no.42 and then it was centrifuged at 5000 rpm for 15 min. Then this filtrate was stored at 5-10^oc for further experiments(Senthilkumar & Sivakumar, 2014).

Synthesis of ZnONPs :

About 0.5 M Zinc sulphate solution was prepared by dissolving 14.38 g of $ZnSO_4$ in 100 ml of distilled water. To this 60 mlof green tea leaf extract was added dropwise until the colour changes from brownishto yellowish colour. It was followed by addition of 0.1 N NaOH dropwise which acts as a stabilizing agent. The precipitate was centrifuged at 8000 rpm for 15 minutes followed by

two times washing with ethanol. Then oven dried at 90^0C for 12 hrs. The dried pelletswerecalcinated in a muffle furnace at 600^0C for 4 hrs to obtain white-coloured zinc oxide nanoparticles(Dhanemozhi et al., 2017; Rajeshkumar et al., 2018).

Characterization techniques:

The Synthesized ZnO NPs were characterized using PSA (ModelHORIBA SZ-100), UV-Vis (Analytik Jena - Specord 210 Plus), XRD (The Empyrean Series III Diffractometer and TEM (Tecnai 12)

Results and Discussion:

The synthesized ZnO NPs were white in colour (Plate 1).The White colour of the nanoparticles is due to biomolecules and polyphenols present in the green tea leaf extract, which covers the surface of the nanoparticles(Fakhari, Jamzad, Kabiri Fard, & reviews, 2019).

Plate 1. Zinc Oxide Nanoparticles

Dynamic light scattering and zeta potential analysis:

The Figure 1 displays the particle size distribution, which was obtained through DLS analysis using the Origin software. The particle size and zeta potential was measured by PSA. The results showed the particle has average particle diameter of 168.8 nm in an aqueous colloidal solution. Additionally, the zeta potential of this particle was 38.2 mV. The result was coincided with result obtained by(Jamdagni, Khatri, & Rana, 2018; Mahendra et al., 2017)

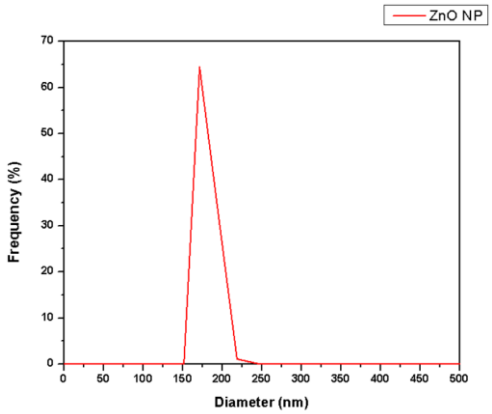

Figure 1. Particle size analysis of biosynthesized zinc oxide nanoparticles

UV – Visible spectrum analysis:

The UV-Vis spectra of ZnO NPs were analysed. It is depicted in Figure 2. The significantly blue-shifted absorption maximum at 323 nm confirms the synthesis of ZnO product ona nano-scale. Typically, for bulk ZnO, the absorption maximum occurs around 380-385 nm. This result was as same as that of the results obtained by(Sutradhar & Saha, 2015; Sutradhar & Saha, 2016). They explained that the reduction in the size of ZnO causes a decrease between two valence bands, thereby causing an increase in frequency which leads to a decrease in wavelength towards the blue end of the spectrum.

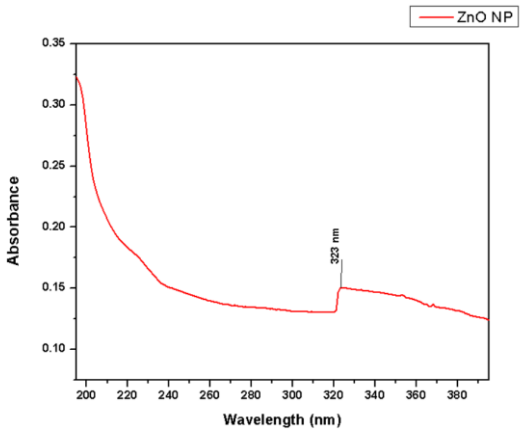

Figure 2. Ultraviolet absorption spectrum of biosynthesized zinc oxide nanoparticles

TEM Analysis:

The morphology of ZnO nanoparticles synthesized was observed using TEM micrograph.The TEM micrograph of the ZnONPs confirmed that theparticles

were almost spherical in shape at the resolution of 100 nm. Energy dispersive X-ray analysis confirmed the presence of zinc and oxygen groups in the sample(Elumalai, Velmurugan, Ravi, Kathiravan, & Ashokkumar, 2015; Viswanathan & Gupta, 2003).

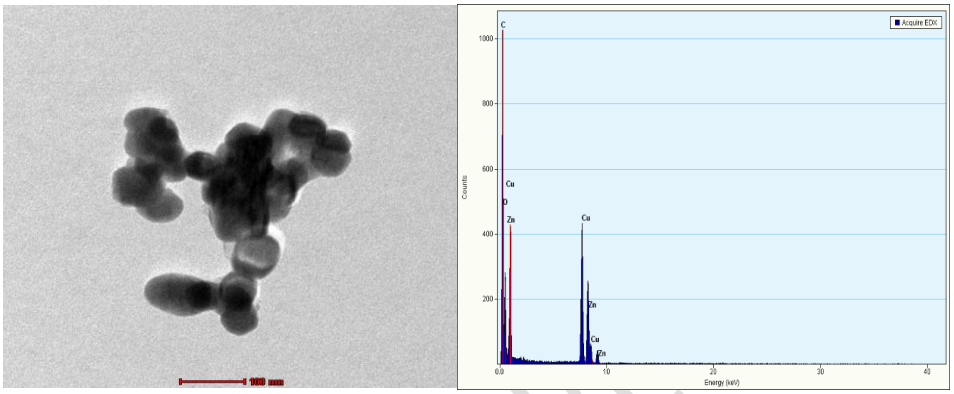

Figure 3. TEM image & EDAX image of biosynthesized zinc oxide nanoparticles

XRD Analysis:

The XRD analysis was done for ZnO NPs. The XRD spectra is given in Figure 5. The ZnO NPs were calcinated before getting XRD spectra. The diffraction peaks that are most prominent are (100), (002), (101), (102), (110), (103), and (112) with diffraction angles of 31.78^0, 34.34^0, 36.18^0, 47.63^0, 56.53^0, 62.79^0, and 67.92^0, respectively. The average particle size (D) of the synthesized nanoparticles was calculated using the Scherrer formula, which is well known. These XRD results were compatible with (Devi & Gayathri, 2014; Irshad et al., 2018; Narendra Kumar et al., 2019) that confirms the prepared ZnO sample is highly crystalline with similar predominant peaks at diffraction angles of 31.78^0, 34.34^0&36.18^0.

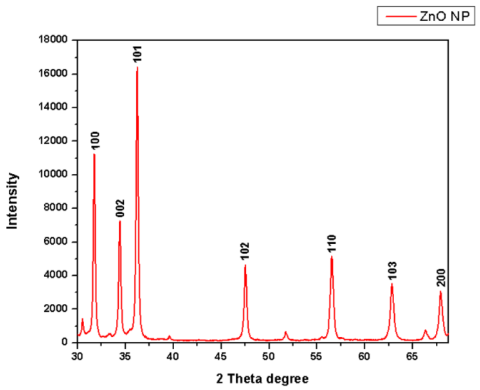

Figure 4. XRD analysis of biosynthesized zinc oxide nanoparticles

Thermogravimetric analysis (TGA):

To determine the effects of high temperature on a specific sample, thermogravimetric analysis is utilized to measure its phase transition, absorption, adsorption, and desorption. In this study,ZnO nanoparticles were analyzed using a temperature program ranging from 27^0C to 900^0C and with a temperature interval of 20^0C per min. The TGA Curve of ZnO NPs is given in Figure 6. The results indicated that at 657^0C, rapid degradation of the zinc oxide nanoparticles takes place, while at 753^0C, the material begins to experience weight loss. This indicates that the biosynthesizedZnOnanoparticles are thermally stable up to 600°Cwhich is similar to (Moharram, Mansour, Hussein, & Rashad, 2014; Rajeshkumar et al., 2018).

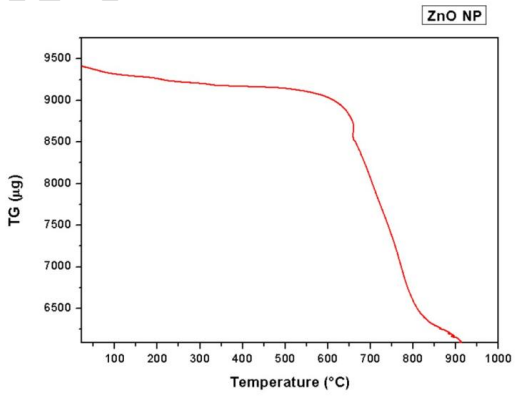

Figure 5. Thermogravimetric analysis of biosynthesized zinc oxide nanoparticles

Conclusion:

The process of using *Camellia sinensis* as a reductant to synthesizeZnO NPs is impressive for its cost-effectiveness, speed, and eco-friendliness. The synthesized ZnO NPs had an average particle diameter of 168.8 nm in an aqueous colloidal solution, with a zeta potential of 38.2 mV. The blue-shifted absorption maximum at 323 nm in the UV-Vis spectrum confirms the particle in is nanoscale. The morphology of the ZnO nanoparticles was observed using TEM micrograph at a resolution of 100 nm, with energy-dispersive X-ray analysis confirming the particle is in spherical shaped and also presence of zinc and oxygen groups in the sample. Calcination at 600°C was done to attain higher crystallinity and to remove all water, as evidenced by the prominent diffraction angles observed at 31.78^0, 34.34^0, 36.18^0, 47.63^0, 56.53^0, 62.79^0, and $67.92^0$with miller indices (100), (002), (101), (102), (110), (103), and (112), respectively in the XRD spectra of the calcined ZnO NPs. The TGA graph of ZnO NPs revealing that rapid degradation of the zinc oxide nanoparticles occurred at 657°C and weight loss began at 753°C indicating that ZnO nanoparticles are thermally stable upto 600^0C.

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